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**RESIN COMPOSITION**

[樹脂組成物]

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(57) [Abstract]

[Constitution]

volume fraction of (A) component which takes continuous phase in resin composition which consists of compound which possesses carboxylic acid group, acid anhydride group, epoxy group, amino group or hydroxy group in the styrenic polymer block of (A) polyamide resin, (B) polyphenylene ether resin, (C) at least two and hydrogenated block copolymer, and modifier (D) intramolecular which consist of olefinic elastomer polymer of at least one, with 44 - 70 percent by volume, (B) with (C) to form dispersed phase, dispersed phase particle diameter being 1.2;  $\mu$ m or less, resin composition. where reduced viscosity ;et r of polyamide in composition molded article is 2.20 (In formic acid, 30°C) or more

[Effect(s) ]

resin composition. where it possessed impact resistance and stiffness in wide temperature range, adjusted heat ageing resistance and had

[Claim(s) ]

[Claim 1]

In resin composition, which consists of compound, which possesses carboxylic acid group, acid anhydride group, epoxy group, amino or hydroxy group in (D) intramolecular as styrenic polymer block of (A) polyamide resin, (B) polyphenylene ether-based resin, (C) at least two and hydrogenated block copolymer, and modifier which consist of olefinic elastomer block of the at

least one,

polyamide of (A) component forms continuous phase, volume fraction with 44 - 70 percent by volume, (B) component and (C) component dispersed phase is formed, dispersed particle diameter is  $1.2\mu\text{m}$  or less, resin composition. which designates that reduced viscosity ;et r of polyamide in composition molded article is 2.20 (In formic acid, 30°C) or more as feature

[Claim 2]

It is a dispersed phase, (B) / resin composition. which is stated in Claim 1 where ratio of (C) component is 80/20 - 50/50 weight ratio

[Claim 3]

resin composition. which is stated in Claim 1, and Claim 2 which add the component (E) stabilizer

[Description of the Invention]

[0001]

[Field of Industrial Application]

this invention is superior in mechanical property, molding property, regards especially impact resistance and thermoplastic resin composition which is superior in heat ageing resistance.

[0002]

[Prior Art]

polyphenylene ether resin, with resin which is superior in dimensional stability, electrical property, heat resistance, water resistance etc, the polystyrene resin and widely is used for industrially in form which blend is done, but large deficiency that it has possessed it is inferior to oil resistance and molding property.

With respect to this, polyamide resin is superior in mechanical strength, oil resistance, heat resistance, etc., is used widely most as representative engineering resin, but it has deficiency that impact resistance at time of heat resistance, drying under dimensional stability, moisture absorption, high load is inferior.

Because of this, it utilizes respective strength of the aforementioned both resins, it can try fact that deficiency of both resins to composition is converted compliment \* with thing as the objective, various composition is proposed so far and is utilized.

composition of this polyphenylene ether resin and polyamide resin is used to show impact strength and the ductility which are necessary for automotive part, because it has possessed the heat resistance which it withstands bake-on painting of online widely, further impact resistance and improvement of heat ageing property are sought, but regarding airbag cover or other application where recently development is advanced.

In Japanese Translation of PCT Publication 63-501580 disclosure inside this, as technology which improves the impact resistance, uses A-B type two block copolymer as rubber component technology which is disclosed, but as for this rubber component it can recognize effect in improvement of impact resistance in low temperature, but rubbery property is lost regarding temperature region which is higher than ambient temperature and there is a defect that it becomes brittle.

On one hand, generality technical (Japan Unexamined Patent Publication Hei 1- 163262 disclosure) which adds copper-based stabilizer which is used is proposed method which adds high-level sterically hindered phenol compound as technology which improves heat ageing resistance, (Japan Unexamined Patent Publication Showa 62-273256 disclosure) and with polyamide.

Example which designates A-B type two block copolymer as rubber component in the Working Example of this Japan Unexamined Patent Publication Hei 1- 163262 disclosure is shown, but defect of fundamental rubber component is not solved, with addition of stabilizer regarding temperature region which is higher than ambient temperature possesses deficiency which degradation is done after all.

Like above furthermore as for material which is superior in heat ageing resistance fact that it is not developed yet is actual condition with the impact resistance in wide temperature range.

[0003]

[Problems to be Solved by the Invention]

As for objective of this invention, in composition which consists of the polyphenylene ether-based resin and polyamide resin, as impact resistance in wide temperature range is granted furthermore with improvement of heat ageing resistance, it is something which aims toward the utilization with field which impact resistance which is represented in the airbag cover, and helmet material, etc., in automotive application from low temperature in broad temperature region to high temperature is required.

[0004]

[Means to Solve the Problems]

Book departure inventors, result of diligent research, impact resistance showing which by fact that it makes specific polyamide resin, polyphenylene ether-based resin and composition which combines specific block copolymer at specific ratio is superior in the wide temperature range, furthermore and, obtaining composition which keeps stiffness reached point of in order to achieve above-mentioned problem.

Furthermore, you discovered fact that improvement of heat ageing resistance is possible by adding stabilizer to this composition, this invention you reached to completion on basis of this knowledge.

[0005]

Namely as for this invention,

1. polyamide of (A) component to form continuous phase in resin composition which consists of compound, which possesses carboxylic acid group, acid anhydride group, epoxy group, amino or hydroxy group in (D) intramolecular as styrenic polymer block of (A) polyamide resin, (B) polyphenylene ether-based resin, (C) at least two and hydrogenated block copolymer, and modifier which consist of olefinic elastomer block of at least one, volume fraction with 44 -70 percent by volume, (B) With (C) forms dispersed phase and dispersed particle diameter is 1.2;μm or less, the resin composition. which designates that reduced viscosity ;et r of polyamide in the composition molded article is 2.20 (In formic acid, 30°C) or more as feature

They are 2.dispersed phase, (B) / description above 1 where ratio of (C) component is 80/20 - 50/50 weight ratio. resin composition. which is stated

3. Furthermore, description above 1 which grants heat ageing resistance by adding (E) stabilizer. 2. resin composition. which is stated

It is something regarding.

[0006]

Below, this invention is explained in detail.

Regarding to this invention, as for polyamide resin which is used as (A) component, being something which possesses amide bond {  $\text{-NH-C(=O)-}$  } in polymer main chain, if it is something which heating and melting it is possible, in each case is a usable.

Making representative ones, 4 -nylon, 6-nylon, 6, 6-nylon, 12-nylon, 6, 10-nylon, terephthalic acid and polyamide, isophthalic acid from hexamethylene diamine and the polyamide, adipic acid from hexamethylene diamine and polyamide, adipic acid and azelaic acid and 2 and 2' -bis from metaxylurene diamine (p-amino cyclohexyl) it can increase polyamide and these copolymerized nylon of polyamide, terephthalic acid and 4, 4' -diamino dicyclohexyl methane ones from -propane.

Among these, 6 -nylon, 6, 6-nylon, 6-6, 6copolymerized nylon sole use or combined use is desirable.

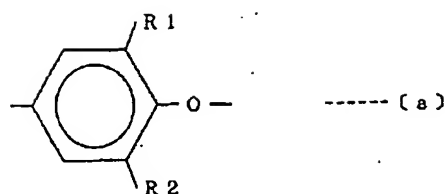
[0007]

Regarding to this invention, polyphenylene ether-based resin which it uses as (B) component, the General Formula which is shown next. [1],

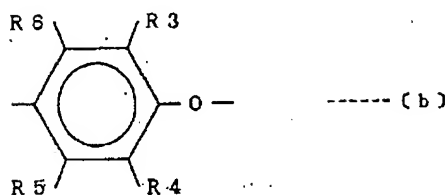
[0008]

[Chemical Formula 1]





(1)



[0009]

It designates (In Formula, as for R1, R2, R3, R4, R5, R6 with residue of alkyl group, aryl group, halogen, hydrogen or other monovalent of the carbon 1-4, as for R5, R6 it is not a hydrogen simultaneously) as repeat unit, constituting unit General Formula [1] [a] and the homopolymer, or copolymer which consists of [b] and these and it can use blend of styrenic polymer.

[0010]

As representative example of homopolymer of polyphenylene ether-based resin, you can list poly (2, 6 -dimethyl -1, 4- phenylene) ether, poly (2 -methyl -6-ethyl 1, 4- phenylene) ether, poly (2, 6 -diethyl -1, 4- phenylene) ether, poly (2 -ethyl -6-n- propyl -1, 4- phenylene) ether, poly (2, 6 -di-n- propyl -1, 4- phenylene) ether, poly (2 -methyl -6-n- butyl -1, 4- phenylene) ether, poly (2 -ethyl -6-isopropyl -1, 4- phenylene) ether, poly (2 -methyl -6-chloroethyl -1, 4- phenylene) ether, poly (2 -methyl -6-hydroxyethyl -1, 4- phenylene) ether, poly (2 -methyl -6-chloroethyl -1, 4- phenylene) ether or other homopolymer.

[0011]

polyphenylene ether copolymer includes polyphenylene ether copolymer which becomes 2, 6 -dimethyl phenol and 2, 3, 6 - trimethyl phenol copolymer of copolymer or o-cresol or 2, 3, 6 - trimethyl phenol and with of o-cresol such as copolymer, polyphenylene ether structure as main component.

[0012]

In addition, if, it does not oppose to gist of this invention in the polyphenylene ether resin of this invention, other various phenylene ether unit where until recently it is proposed that it is possible to exist in polyphenylene ether resin, including, as the partial structure it does not care.

It is stated in Japan Patent Application Sho 63-12698 number and Japan Unexamined Patent Publication Showa 63-301222 disclosure trace as example of those where it is proposed that it coexists, 2 - (dialkyl aminomethyl) - you can list 6-methyl phenylene ether unit and 2 - (N- alkyl -N- phenyl amino methyl) - 6 - methyl phenylene ether unit etc.

[0013]

In addition, diphenoquinone, etc., trace those which are connected is included in main chain of polyphenylene ether resin.

As for polyphenylene ether resin which is used for this invention, 0.30 - 1.5, those of range of preferably 0.35-1.0 are ideal with inherent viscosity (chloroform solution, 30°C) .

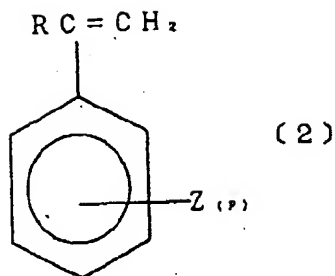
[0014]

In addition, regarding to this invention, styrenic resin which polyphenylene ether resin is jointly used, polymerizing styrenic compound, styrenic compound and copolymerizable compound under rubber polymer existing or absence, is polymer which is acquired.

styrenic compound, General Formula [2]

[0015]

[Chemical Formula 2]



[0016]

compound which is displayed with (In Formula, R shows hydrogen, lower alkyl or halogen, Z is selected from group which consists of vinyl, hydrogen, halogen and lower alkyl, the p is integer 0 - 5) is meant.

As these embodiment, you can list styrene, a -methylstyrene, 2, 4- dimethyl styrene, monochlorostyrene, p- methylstyrene, p- t- butyl styrene, ethyl styrene etc.

In addition, "methyl methacrylate, ethyl methacrylate or other methacrylic acid ester", such as "acrylonitrile, methacrylonitrile or other unsaturated nitrile compound" and maleic anhydride or other acid anhydride are listed as styrenic compound and copolymerizable compound, with styrenic compound are used.

In addition, copolymer of conjugated diene rubber or conjugated diene and aromatic vinyl compound and you can list these hydrogenation product or ethylene -propylene copolymeric rubber, etc., as rubber polymer.

[0017]

manufacturing method of styrenic resin of this invention is not something which is limited, block polymerization which is well known in person skilled in the art, making use of which of solution polymerization, emulsion polymerization, suspension polymerization it is good.

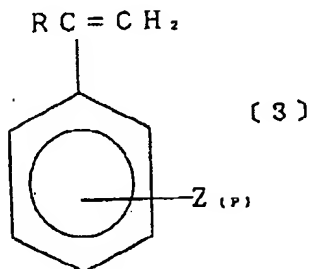
Regarding to this invention, hydrogenated block copolymer which is a (C) component is block copolymer which consists of with styrenic polymer block of at least two and olefinic elastomer block of the at least one.

[0018]

styrenic polymer block which is said to this invention concrete  
General Formula [3]

[0019]

[Chemical Formula 3]



[0020]

It is a polymer or a copolymer block which is induced from compound which is displayed with (In Formula, R shows hydrogen, lower alkyl or halogen, Z is selected from group which consists of vinyl, hydrogen, halogen and lower alkyl, the p is integer 0 - 5) .

olefinic elastomer block which is said to this invention, with polymer block which possesses form which olefin compound of one kind or more which is chosen from inside the ethylene, propylene, 1- butene, isobutylene or other monoolefin or butadiene, isoprene, 1, 3- pentadiene or other conjugated diolefin, 1, 4- hexadiene, norbornene derivative or other unconjugated diolefin polymerizes or copolymerizes, or, furthermore as for degree of unsaturation of said block it is 20% or less.

Therefore, as constituent monomer of olefinic elastomer block, when above-mentioned diolefin are used, treatment which decreases degree of unsaturation with hydrogenation, etc., to extent where degree of unsaturation of said block part amount does not exceed 20% must be administered.

To also, olefinic elastomer block styrenic compound to random may be copolymerized.

One where molecular weight is high in regard to content of hydrogenated block copolymer is desirable, in addition ratio of

styrene /rubber being styrene amount, combination of same 15 - 40 weight % as those of 50 - 80 weight % improvement of impact resistance, especially, impact property after heat ageing is desirable highly.

[0021]

As for proportion of resin component in this invention, polyamide of (A) component forms continuous phase and (B) with (C) forms dispersed phase, it is desirable for volume fraction of (A) to be range of 44 - 70 percent by volume.

When volume fraction is not full in 44%, surface external appearance of molded article becomes the deficiency, when it exceeds 70%, because balance of heat resistance and stiffness changes, is not desirable.

It is a dispersed phase next, (B) with ratio of (C) range 80/20 - 50/50 is desirable.

When (B) component exceeds 80%, impact resistance becomes insufficient, when it is less than 50%, in order heat resistance and stiffness greatly to decrease, is not desirable.

[0022]

Regarding to this invention, compound which is used possesses carboxylic acid group, acid anhydride group, epoxy group, amino group or hydroxy group simultaneously in intramolecular as (D) component, is unsaturated dicarboxylic acid and its derivative, and the hydroxycarboxylic acid.

maleic acid, fumaric acid, chloro maleic acid, cis- 4 - cyclohexene -1, 2 dicarbonic acid and these acid anhydride, ester, semi- alkyl ester, amide, imide, etc., and you can list citric acid, lactic acid, malic acid etc, but especially, a, b- unsaturated dicarboxylic acid and its derivative, concretely, citric acid, fumaric acid, maleic acid and maleic anhydride being ideal, the maleic anhydride is most desirable.

It is good using unsaturated dicarboxylic acid and its derivative and hydroxycarboxylic acid, respectively with alone it is good using and combining 2 kinds or more.

As for addition quantity of (D) component, it is desirable to choose in range of 0.05 - 5 parts by weight, preferably 0.1 ~3 parts by weight, description above (A), with respect to total

amount 100parts by weight of (B) and (C) component.

When addition quantity, under 0.05 parts by weight, particle diameter of dispersed phase becomes large and on property is not desirable, in addition exceeds 5 parts by weight there is a problem such as coloration of resin and decrease of property, is not desirable.

[0023]

stabilizer which is used for (E) component of this invention is used for the stabilizer and (B) polyphenylene ether-based resin of (A) polyamide resin and stabilizer of rubber component.

As stabilizer of (A) polyamide resin, besides it can use amines, hindered phenols and the phosphite ester which are a stabilizer of general resin, halide of copper or nickel, tin or other metal compound and the alkali metal and combined use of both are known.

In large revised objective of heat resistance it is general to be able to use the latter.

[0024]

As metal compound, it can increase copper salt or copper chelate compound of organic acid a metal salt of inorganic acid like copper (I) chloride, copper (II) chloride, copper iodide, copper sulfate, copper nitrate, copper phosphate and like the copper acetate, copper salicylate, copper stearate, copper benzoate, lactic acid copper, sebacic acid copper, but preferred is copper acetate, copper iodide.

As halide of alkali metal, it can increase potassium iodide, potassium bromide, potassium chloride, sodium iodide, sodium bromide, sodium chloride, but preferred is the potassium iodide.

As for polyamide resin which is stabilized with this copper compound and halide, etc., of alkali metal in order easily with obtainable, to obtain composition of this invention with market, effective amount remains and others using commercial polyamide is desirable.

In addition regarding to this invention, being important to be omnipresent in polyamide resin, beforehand it uses copper-based stabilizer with stabilizer of high concentration or as master batch, or it is desirable to use polyamide which includes copper

of necessary quantity in final composition.

[0025]

As for amount used of stabilizer of polyamide resin which is used for the this invention, as for amines, hindered phenols and phosphite ester they are 0.01 - 2 parts by weight, preferably 0.05~1 part by weight with respect to polyamide resin 100parts by weight which is used.

As for metal compound in same way as for halide of 0.0005 - 0.2 parts by weight, preferably 0.001~0.1 parts by weight, alkali metal it is used in range of 0.005 - 1.0 parts by weight, preferably 0.02~0.5parts by weight as metal component.

As (B) polyphenylene ether-based resin and stabilizer of rubber component, it can use stabilizer and its combination which are shown below.

[0026]

1.metal sulfide and combination 2.metal inactivator of metal oxide

3.amines

4.hindered phenols

5.phosphite ester

6.thioether

When these are explained concretely,

As 1.metal sulfide and combination of metal oxide, of zinc sulfide and combination with zinc oxide or magnesium oxide,

As 2. metal inactivator, adipic acid dihydrazide, isophthalic acid dihydrazide or other hydrazine derivative, Irganox (trademark) MD 1024 (Ciba-Geigy Japan Limited make), melamine and its derivative or the analogous compound (compound which is stated in Japan Unexamined Patent Publication Hei 4- 239547 disclosure, Japan Unexamined Patent Publication Hei 4- 239556 disclosure, Japan Unexamined Patent Publication Hei 4- 239557 disclosure and Japan Unexamined Patent Publication Hei 4- 239558 disclosure each disclosure) and Adekastab (trademark) mark CDA-1, mark CDA-6, mark ZS-27, mark ZS-81 (Or more Asahi Denka Kogyo

K.K. (DB 69-057-1187) make), Stabinol (trademark) CS-42 (Sumitomo Chemical Co. Ltd. (DB 69-053-5307) make), etc.,

It can use compound which is stated in Japan Unexamined Patent Publication Hei 4- 239556 disclosure 3. amines, 4. hindered phenols, 5. phosphite ester and as 6. thioether.

[0027]

As for amount used of stabilizer of resin which consists of polyphenylene ether-based resin and rubber polymer which are used for this invention, range of 0.01 - 5 parts by weight is desirable with respect to resin 100 parts by weight, furthermore 0.05 - 2 parts by weight are more desirable.

0.01 Under parts by weight improvement effect not to be a fully, exceeding 5 parts by weight, adding, improvement effect is small, also decrease of heat resistance and impact resistance is recognized and is not desirable.

[0028]

As for stabilizer of resin which consists of this polyphenylene ether-based resin and the rubber polymer in order selectively to distribute to said resin, it is desirable to combine, for that it is desirable to knead beforehand, but As for composition of polyamide resin and polyphenylene ether-based resin step which modified is done being necessary with modifier, effect in order to decrease considerably, is not desirable with this step including the stabilizer.

Because of this you can think method which adds stabilizer after modified, but if only it adds simply, selectively it is not possible to distribute and, there is a problem that effect is small.

method which kneaded stabilizer with such as polyphenylene ether-based resin, polystyrene resin and rubber polymer with the high concentration as this response, adds in form of so-called master batch being effective, most is method which has practicality then.

[0029]

configuration of this invention temperature and time method which each component which is done melt mixing are done if it sets with types and the proportion of resin component which is



used, are good, but usually temperature of 240 - 360°C, preferably 260-340°C, in addition kneading time of 0.1 - 10 min, preferably 0.3-3 min extent is suitable.

As melt mixing device, it can use single screw extruder, twin screw extruder, heated roll, Banbury mixer, kneader etc, but extruder is desirable, twin screw extruder is most desirable even among them.

[0030]

According to desire other additive, for example, glass fiber or carbon fiber and reinforcing agent, or particle, fibrous filler, flame retardant, plasticizer, ultraviolet absorber, colorant, mold release, etc., like various whisker can be added in composition of this invention.

[0031]

[Working Example(s) ]

Next, this invention is explained concretely with Working Example.

Working Example below none with illustrate ones, is something which limits content of this invention.

component which is used in Working Example and Comparative Example is something below.

(A) polyamide resin

With A-1:  $hr = 3.0$  (98% sulfuric acid, 25°C), amino end group concentration being  $4.4 \times 10^{-5}$  mole/g, end carboxyl concentration the polyamide 6. of  $5.5 \times 10^{-5}$  mole/g

(density  $1.14 \text{ g/cm}^3$ )

With A-2:  $hr = 3.2$  (98% sulfuric acid, 25°C), amino end group concentration being  $4.3 \times 10^{-5}$  mole/g, end carboxyl concentration the polyamide 6. of  $4.9 \times 10^{-5}$  mole/g

(density  $1.14 \text{ g/cm}^3$ )

With A-3:  $hr = 2.8$  (98% sulfuric acid, 25°C), amino end group concentration being  $4.5 \times 10^{-5}$  mole/g, end carboxyl concentration the polyamide 6. of  $6.0 \times 10^{-5}$  mole/g

(density 1.14 g/cm<sup>3</sup>)

With A-4: hr = 2.5 (98% sulfuric acid, 25°C), amino end group concentration being  $4.3 \times 10^{-5}$  mole/g, end carboxyl concentration the polyamide 6. of  $7.2 \times 10^{-5}$  mole/g

(density 1.14 g/cm<sup>3</sup>)

With A-5: hr = 2.3 (98% sulfuric acid, 25°C), amino end group concentration being  $3.6 \times 10^{-5}$  mole/g, end carboxyl concentration the polyamide 6. of  $8.6 \times 10^{-5}$  mole/g

(density 1.14 g/cm<sup>3</sup>)

With A-6: hr = 3.0 (98% sulfuric acid, 25°C), amino end group concentration being  $7.5 \times 10^{-5}$  mole/g, end carboxyl concentration the polyamide 6. of  $3.1 \times 10^{-5}$  mole/g

(density 1.14 g/cm<sup>3</sup>)

With A-5: hr = 2.5 (98% sulfuric acid, 25°C), amino end group concentration being  $7.7 \times 10^{-5}$  mole/g, end carboxyl concentration the polyamide 6. of  $4.5 \times 10^{-5}$  mole/g

(density 1.14 g/cm<sup>3</sup>)

polyamide above other than those which especially are clearly written used those which are controlled within water content 500ppm.

(B) polyphenylene ether resin

poly (2 and 6 -dimethyl -1, 4- phenylene) ether. where B-1: inherent viscosity is 0.52 dl/g (In 30°C, chloroform)

(density 1.06 g/cm<sup>3</sup>)

(C) hydrogenated block copolymer

C-1: styrene -ethylene /butylene -styrene -ethylene /butylene four type block copolymer ( $M_n = 12 \times 10^4$ , styrene polymer block /olefin polymer block = 35/65weight ratio, oil component 35 weight % contents and density 0.90 g/cm<sup>3</sup>)

C-2: styrene -ethylene /butylene -styrene three type block copolymer ( $M_n =$  approximately  $15 \times 10^4$ , styrene polymer block

/olefin polymer block ratio = approximately 33/67 weight ratio, density = 0.92 g/cm<sup>3</sup>)

C-3: styrene -ethylene /butylene -styrene -ethylene /butylene four type block copolymer ( $M_n = 7 \times 10^4$ , styrene polymer block /olefin polymer block = 30/70 weight ratio, density 0.91 g/cm<sup>3</sup>)

C-4: styrene -ethylene /butylene -styrene -ethylene /butylene four type block copolymer ( $M_n = 9 \times 10^4$ , styrene polymer block /olefin polymer block = 60/40 weight ratio, density 0.97 g/cm<sup>3</sup>)

C-5: styrene -ethylene /propylene two type block copolymer ( $M_n =$  approximately  $10 \times 10^4$ , styrene polymer block /olefin polymer block = approximately 37/63 weight ratio, density = 0.92 g/cm<sup>3</sup>)

compound which possesses carboxylic acid group, acid anhydride group, epoxy group, amino group or hydroxy group simultaneously in (D) intramolecular

D-1: maleic anhydride

polyamide which is stabilized with stabilizer, said stabilizer of (E) polyamide

With polyamide 6 which in order with calculation as copper to become 0.1 weight %, combines copper (I) iodide to E-1: polyamide 6, melt mixing it does, hr = 2.8 (98% sulfuric acid, 25°C)

[0032]

E-2: B-1 60 parts by weight, Ciba-Geigy make Irganox (trademark) 1076 15 parts by weight, Ciba-Geigy make Irgafos (trademark) 168 after 10 parts by weight blending, melt mixing doing 15 parts by weight, Ciba-Geigy make Irganox (trademark) MD 1024 with laboratory plastic mill, powder fragment it did, made master batch of stabilizer.

[0033]

property evaluation condition and method are as follows.

test piece was produced with injection molding machine (Toshiba Machine Co. Ltd. (DB 69-055-0983) make making use of IS80C, cylinder temperature 280°C, molding cycle 1 min), following property measurement and test were executed.

(1) MFR (melt flow rate)

It conforms to JIS K-7210, measures with 280°C, load 5 kg.

(2) heat distortion temperature
4
4
It conforms to ASTM D648
6 K
6 K
It measures with G load.

thickness)

It conforms to ASTM D256, 23°C - measures with 30°C.

Quantity of (4) surface impact ductile break

Making use of graphic impact tester {Toyo Seiki Ltd. supplied}, 3 mm platelet were done 5 test, number of layers which ductile break is done was sought.

It did measurement temperature, with 100°C, 23°C, -30°C.

(5) tensile strength, elongation

Conforming to ASTM -D638, it sought.

(6) flexural strength, flexural modulus

Conforming to ASTM -D790, it sought.

(7) heat ageing test

a. Izod impact strength

Izod piece of 1/8 inch it tested in oven of 120°C, 500hours, started taking aging in 1000 hours, with 23°C - 30°C measured with approximately aging retention sought.

measurement method conforms to (3) .

[0034]

Quantity of b. surface impact ductile break

test piece of thickness 3 mm it tested in oven of 120°C, 500 hours, started taking aging in 1000 hours, with 100°C, 23°C and - 30°C measured, number of layers which ductile break is done it sought.

It conforms to (4) .

(8) dispersed particle diameter

composition was measured after melting, making use of laser diffraction type particle size distribution measurement device (SALD2000: Shimadzu Corporation (DB 69-055-8747) make) to formic acid, 50% cumulative particle diameter was sought.

(9) polyamide hr

After melting molded article which is used for physical measurement in formic acid, after separating dispersed phase due to centrifugal separation, reprecipitation after doing, vacuum drying was done with water, polyamide was removed.

Next, reduced viscosity hr was measured with 30°C in formic acid making use of Canon Inc. (DB 69-054-9662) Fenske type viscometer tube.

(Furthermore formic acid which is used special grade reagent of purity 99%)

[0035]

[Working Example 1]

polyphenylene ether-based resin (B-1) 34 parts by weight and rubber polymer (C-1) 16 parts by weight and modifier (D-1) the top of corotating twin screw extruder of screw diameter 25 mm (prestage) from it supplied at ratio of 0.3 parts by weight, supplying 50 parts by weight from extruder halfway (midstage), extrusion it mixed polyamide 6 (A-1) with 300°C, 300rpm, sprayed amount 20 kg/hr and acquired composition pellet.

Next it tested variously before with method which was inscribed.

evaluation result is shown in Table 1.

[0036]

Other than modifying in those which show {Working Example 2 ~3} (A) component in Table 1 it executed completely in same way as Working Example 1.

Result is shown in Table 1.

[0037]

Other than modifying in those which show {Comparative Example 1~2} (A) component in Table 1 it executed completely in same way as Working Example 1.

Result is shown in Table 1.

[0038]

[Comparative Example 3]

addition quantity of (D) component increased weight other than doing, it executed completely in same way as Working Example 3 in 0.6 part.

Result is shown in Table 1.

[0039]

[Comparative Example 4]

Other than modifying extruder temperature in 330°C, it executed completely in same way as Working Example 3.

Result is shown in Table 1.

[0040]

[Comparative Example 5]

Adjusting polyamide of (A-3) component water content 2000 ppm, other than using, it executed completely in same way as Working Example 3.

Result is shown in Table 1.

[0041]

[Comparative Example 6]

Other than modifying in those which show (C) component in Table 1 it executed completely in same way as Working Example 1.

Result is shown in Table 1.

[0042]

{Working Example 4~6} (A) (B) And other than modifying in quantity which shows (C) component in Table 2 it executed completely in same way as the Working Example 1.

Result is shown in Table 2.

[0043]

[Working Example 7]

Other than modifying in those which show (C) component in Table 2 it executed completely in same way as Working Example 5.

Result is shown in Table 2.

[0044]

[Working Example 8]

Other than modifying in those which show (A) component in Table 2 it executed completely in same way as Working Example 5.

Result is shown in Table 2.

[0045]

Other than modifying in quantity which shows {Comparative Example 7~9} (A) component in Table 2 it executed completely in same way as Working Example 4~6.

Result is shown in Table 2.

[0046]

[Comparative Example 10]

Other than modifying in those which show (A) component in Table 2 it executed completely in same way as Working Example 7.

Result is shown in Table 2.

[0047]

[Comparative Example 11]

Other than modifying in those which show (A) component in Table 2 it executed completely in same way as Working Example 8.

Result is shown in Table 2.

[0048]

Other than modifying in those which show {Working Example 9-10} (C) component in Table 2 it executed completely in same way as Working Example 5.

Result is shown in Table 2.

[0049]

[Working Example 11]

polyphenylene ether-based resin (B-1) 34 parts by weight and rubber polymer (C-1) 16 parts by weight and modifier (D-1) the top of corotating twin screw extruder of screw diameter 25mm (prestage) from it supplied at ratio of 0.3 parts by weight, polyamide 6 (A-1) supplying 5 parts by weight and stabilizer (E-2) from 1 part by weight, extruder halfway (midstage), extrusion it mixed 45 parts by weight, stabilizer-containing polyamide (E-1) with 300°C, 300rpm, sprayed amount 20kg/hr and acquired composition pellet.

Next it tested variously before with method which was inscribed.

evaluation result is shown in Table 3.

[0050]

Other than modifying in those which show {Working Example 12-13} (A) component in Table 3 it executed completely in same way as Working Example 11.



Result is shown in Table 3.

[0051]

Other than modifying in those which show {Comparative Example 12~13} (A) component in Table 3 it executed completely in same way as Working Example 11.

Result is shown in Table 3.

[0052]

[Comparative Example 14]

Other than modifying in those which show (C) component in Table 3 the Working Example 11 it executed completely in same way.

Result is shown in Table 3.

[0053]

Other than modifying in {Working Example 14~16} (B) component and quantity which shows (C) component in Table 3 it executed completely in same way as Working Example 11.

Result is shown in Table 3.

[0054]

[Working Example 17]

Other than modifying in those which show (A) component in Table 3 it executed completely in same way as Working Example 15.

Result is shown in Table 3.

[0055]

Other than modifying in those which show {Working Example 18~19} (C) component in Table 3 it executed completely in same way as Working Example 15.

Result is shown in Table 3.

[0056]

Other than modifying in those which show {Comparative Example

15-16} (A) component in Table 3 it executed completely in same way as Working Example 15.

Result is shown in Table 3.

[0057]

[Table 1]

1. Working example 1
2. Working example 2
3. Working example 3
4. Comparative example 1
5. Comparative example 2
6. Comparative example 3
7. Comparative example 4
8. Comparative example 5
9. Comparative example 6
10. Composition (percent by weight)
11. Polyamide resin
12. Polyphenylene ether resin
13. Rubberized polymer
14. Denaturant
15. Stabilizer
16. Analysis
17. Dispersed particle diameter
18. PA  $\eta_r$  in formed product
19. Dissolved polyamide rate
20. Physical characteristics
21. Thermoplastic temperature
22. Izod impact strength
23. Tensile strength
24. Tensile elongation
25. Bending strength
26. Bending tensile strength

14. Denaturant
15. Stabilizer
16. Analysis
17. Dispersed particle diameter
18. PA  $\eta_r$  in formed product
19. Dissolved polyamide rate
20. Physical characteristics
21. Thermoplastic temperature
22. Izod impact strength
23. Tensile strength
24. Tensile elongation
25. Bending strength
26. Bending tensile strength

		実施例 -4	実施例 -5	実施例 -6	実施例 -7	実施例 -8	比較例 -7	比較例 -8	比較例 -9	比較例 -10	比較例 -11	実施例 -9	実施例 -10
組成 (重量%)	ポリマイト 樹脂	50	50	50	50							50	50
	ポリマイト 樹脂 8-1	32	30	28	30	30	32	30	28	30	30	30	30
	エポキシ重合体	18	20	22	20		18	20	22	20		15	10
	C-1											5	5
	C-2											5	5
	C-3												
	C-4												
安定剤	C-5												
	D-1	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3
	E-1												
解析	E-2												
	分散粒径 μm	0.80	0.95	0.91	0.98	0.85	1.25	1.45	1.62	1.51	1.45	0.91	0.94
	成形品中 PA 率	2.35	2.44	2.41	2.41	2.45	2.08	2.11	2.10	2.06	2.08	2.41	2.45
	ポリマイト 容積分率 Vol%	47	46	46	47	46	47	46	46	47	46	47	47
物性	MFR g/10min	13	18	18	11	5	26	22	24	14	9	18	21
	熱変形温度 ℃	166	160	163	165	165	157	155	159	163	161	163	163
	70℃ 引張強度 Kg・cm/cm												
	23℃ 引張強度 Kg・cm/cm	105	108	112	102	102	74	78	80	81	77	118	121
物性	-30℃ 引張強度 Kg・cm/cm	81	86	90	85	83	40	45	51	49	49	96	100
	面衝撃破壊枚数 枚	5	5	5	5	5	5	5	5	5	5	5	5
	100℃ 引張強度 Kg/cm <sup>2</sup>	5	5	5	5	5	5	5	5	5	5	5	5
	23℃ 引張強度 Kg/cm <sup>2</sup>	5	5	5	5	5	3	4	5	5	5	5	5
物性	-30℃ 引張強度 Kg/cm <sup>2</sup>	5	5	5	5	5	1	2	3	2	2	5	5
	引張強度 Kg/cm <sup>2</sup>	500	490	450	510	520	450	420	390	410	430	500	500
	引張伸度 %	130	140	130	120	120	100	110	100	100	110	160	160
	曲げ強度 Kg/cm <sup>2</sup>	690	600	570	660	680	650	580	550	550	580	600	610
物性	曲げ弾性率 Kg/cm <sup>2</sup>	17200	15300	15000	17100	17500	16300	14100	13800	14200	14900	15900	16000

[0059]

[Table 3]

1. Working example 11
2. Working example 12
3. Working example 13
4. Comparative example 12
5. Comparative example 13
6. Comparative example 14
7. Working example 14
8. Working example 15
9. Working example 16
10. Working example 17
11. Working example 18
12. Working example 19
13. Comparative example 15
14. Comparative example 16
15. Composition (percent by weight)
16. Polyamide resin
17. Polyphenylene ether resin
18. Rubberized polymer
19. Denaturant
20. Stabilizer
21. Analysis
22. Dispersed particle diameter
23. PA  $\eta_r$  in formed product
24. Dissolved polyamide rate
25. Physical characteristics
26. Izod impact strength
27. Sheets broken by surface impact
28. Izod impact strength preservation rate
29. Sheets bent by surface impact

	実施例 -11	実施例 -12	実施例 -13	比較例 -12	比較例 -13	比較例 -14	比較例 -14	実施例 -15	実施例 -16	実施例 -17	実施例 -18	実施例 -19	比較例 -15	比較例 -16
組成 (質量%)														
ポリミド樹脂	45					45	45	45	45		45	45		
A-1		45		45									45	
A-2														
A-3														
A-4														
A-5														
A-6														
A-7										45				
ポリエーテル樹脂 B-1	34	34	34	34	34	34	34	30	28	30	30	30	30	45
ガラス状重合体	16	16	16	16	16		18	20	22	20	15	10	20	20
C-1														
C-2														
C-3														
C-4														
C-5														
硬化剤	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3
D-1														
E-1	5	5	5	5	5	5	5	5	5	5	5	5	5	5
E-2	1	1	1	1	1	1	1	1	1	1	1	1	1	1
分散剤	0.61	0.57	0.66	1.30	1.30	0.64	0.83	0.97	0.90	0.82	0.94	0.97	1.45	1.33
成形品中 P A 77	2.40	2.52	2.27	2.12	1.95	2.44	2.41	2.40	2.49	2.42	2.46	2.50	2.09	2.11
折れ率	47	47	47	47	47	47	47	46	46	47	46	46	47	47
777 外衝撃強度 Kg-cm/cm														
23 °C	102	102	98	94	74	81	104	107	113	105	116	121	77	79
-30 °C	72	76	69	54	35	73	83	87	90	85	95	99	49	53
面衝撃強度														
100 °C	5	5	5	5	5	3	5	5	5	5	5	5	5	5
23 °C	5	5	5	5	3	5	5	5	5	5	5	5	4	4
-30 °C	5	5	5	3	1	5	5	5	5	5	5	5	1	2
777 外衝撃強度保持率 %														
0 Hrs	100	100	100	100	100	100	100	100	100	100	100	100	100	100
500 "	83	85	75	45	25	70	84	89	92	95	93	94	51	46
1000 "	45	48	40	25	21	35	53	55	57	61	73	75	28	30
面衝撃強度破壊枚数														
-30 °C	5	5	5	3	1	5	5	5	5	5	5	5	4	3
0 Hrs	5	5	5	2	0	4	5	5	5	5	5	5	1	2
500 "	5	5	5	0	0	2	5	5	5	5	5	5	0	0
1000 "	5	5	5	5	5	3	5	5	5	5	5	5	5	5
100 °C	5	5	5	5	5	1	5	5	5	5	5	5	5	5
0 Hrs	5	5	5	5	5	1	5	5	5	5	5	5	5	5
500 "	5	5	5	5	5	0	5	5	5	5	5	5	5	5
1000 "	5	5	5	3	0	0	5	5	5	5	5	5	4	4

[0060]

[Effects of the Invention]

resin composition of this invention set reduced viscosity hr and dispersed phase particle diameter of the polyamide in composition molded article to a certain specific range, material which had impact resistance and stiffness in wide temperature range furthermore by limiting proportion of each component, acquired.

Furthermore development to field which impact resistance where it adjusts heat ageing resistance which is superior and by adding heat stabilizer to this material, it becomes material which it had, is represented in airbag cover, and the helmet material, etc., in automotive application from low temperature in broad temperature region to high temperature is required became possible.

**Szekely, Peter**

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**From:** Releford, Carol  
**Sent:** Monday, August 29, 2005 1:45 PM  
**To:** Szekely, Peter  
**Subject:** Re: Copy of Machine Translations - 08-217972.jaa (S/N 10/509,612)



08-217972  
.jaa.doc

Hi Peter,

Here's a copy of the Machine Translation that you requested. For S/N 10/509,612.

tx